CELLULAR PROFILES IN DIRECTIONAL SOLIDIFICATION: IS THE SAFFMAN-TAYLOR BRANCH OF SOLUTIONS THE PHYSICALLY RELEVANT ONE?

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ABSTRACT

We summarize the main results and implications of our work on the calculation of cellular shapes in directional solidification using the asymptotic matching method introduced by Dombre and Hakim. For cells with narrow grooves, the finite Péclet number corrections to the cellular profiles that reduce to the Saffman-Taylor solutions for Péclet number $p \to 0$, turn out to be small. We argue that there are several discrepancies between the behavior of these Saffman-Taylor like cells and those observed in experiments as well as numerical studies that suggest that this branch of solutions is not always the physically relevant one for directional solidification.

INTRODUCTION

In the last few years, most analytic approaches[1-5] aimed at understanding the problem of directional solidification (DS) have been based on the observation (originally due to Pelcé and Pumir[1]) that the directional solidification equations for the two-dimensional one-sided model can be reduced to those for the Saffman-Taylor (ST) problem in the small Péclet number limit $p \to 0$. With this mapping one can therefore easily obtain explicit results[3,4] for a continuous branch of cellular shapes with deep grooves.

For cells with narrow grooves, Dombre and Hakim[2] showed that one can exploit the smallness of the groove width for the calculation of cell parameters using asymptotic matching methods. For the limit $p \to 0$ they studied, their results reduce to those derived directly from the mapping onto the ST problem. Motivated by the observation that experimental cells often have rather narrow grooves, we have extended[6,7] their treatment of cells with narrow grooves to the case of small partition coefficient k ($k \leq 0.15$, say) but p of order unity. This is the range relevant for many experiments. Our results reduce smoothly to those of Dombre and Hakim[2] for $p \to 0$, where the

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restriction to small k is then not required, and we will therefor refer to this particular branch of solutions we calculate as the Saffman-Taylor-like branch.

These results for cells with narrow grooves are disappointing in that they do not compare well with typical experimental results. Moreover, they also show discrepancies with a number of numerical studies of the same two-dimensional one-sided model. They therefore raise the question whether this branch of solutions is the experimentally relevant one, and also whether this is in branch of solutions studied in most numerical approaches.[8–10]

The essential problem is easy to understand qualitatively for $p \to 0$. It is well-known that the appropriate dimensionless surface tension parameter σ of experimental cells usually turns out to be rather small, of the order of 10^{-2}, and that cells typically have rather *narrow* grooves. This behavior is incompatible with that of the ST branch of solutions, for which small surface tension cells correspond to *wide* groove solutions: in the absence of surface tension anisotropy these solutions have grooves whose width not too far from the tip is roughly half of the pattern wavelength λ (See, e.g., Refs 3 and 4).

This discrepancy is just one of the ways in which ST type of cellular solutions appear to behave significantly different from experimental cells as well as from most cellular solutions studied numerically for the same model. We have recently given an extensive discussion of these issues,[7] and therefore confine ourselves here to highlighting some of our main points, in particular those that touch on other topics addressed at the summerschool.

SUMMARY OF RESULTS AND IMPLICATIONS

In this section, we briefly summarize the main results and conclusions from our analysis of cellular profiles with narrow grooves using matched asymptotic expansions.[7] We then illustrate these points in some more detail in the subsequent sections. Throughout this paper, we will denote the relative groove width of cells near their tip by ε, since this is the small parameter in the matching method. Note that this quantity is usually referred to as $1-\lambda$ in the ST literature. However, we prefer to use λ for the wavelength.

1. For systems with small k, we can calculate the properties of a particular continuous branch of steady state solutions describing cellular profiles with deep narrow grooves. The Péclet number dependence of these solutions is weak, and for $p \to 0$ they reduce to the Saffman-Taylor-like solutions found earlier by Dombre and Hakim[2]. The variation in wavelength of this branch is parametrized in the matching theory by the relative groove width ε near the tip, or, more convenient to experiment, by a dimensionless parameter ζ_t giving the tip position (see the next section and Eq. (3) below).

2. A fundamental assumption self-consistently satisfied by these solutions is the *existence of a single matching region controlled by the small parameter* ε *where* deep narrow grooves can be matched to a particular class of finite amplitude cellular solutions describing the fingertip region. These finite amplitude solutions have wavelength lying *outside* the planar instability band and evolve from the (infinitesimal amplitude) neutrally stable modes forming the short wavelength side of the planar instability band (the left hand solid curve in Fig. 1; see below). These solutions have a regular expansion in powers of k. The fingerlike solutions with narrow grooves (small ε) resulting from the matching also have wavelengths lying outside the planar instability band and have tips that do not move up nearly as much as one expects[11] on the basis of a simple extension of

the classical constitutional supercooling argument (the Local Constitutional Supercooling criterion or LCS – see the next section).

3. Most numerical studies, using the same two-dimensional model we consider here, find narrow-grooved fingerlike patterns, but with wavelength lying *inside* the planar instability band. These solutions are found well above threshold and evolve from small amplitude cells with wavelengths lying inside the planar instability band, presumably related for small k to the cells studied by Sivashinsky[12] and others.[13,14] The latter bifurcate off from the *center* of the planar instability band (indicated by the square in Fig. 1), and do not have a regular k expansion. The contrast with the matching solutions discussed in 1) is evident.

4. Most experimental patterns also have narrow grooves, but with wavelength lying inside the planar instability band. In the few cases where experimental data on the tip position is available, small values of the parameter ζ_t are found, in qualitative agreement with our approximate stability criterion[11] (LCS). This implies that the tips of experimentally selected patterns have moved up substantially in the temperature gradient relative to the planar position.

5. In contrast, the matching solutions for small ε have tip positions that remain relatively close to the planar position. More detailed experimental measurements of the tip position would be extremely useful to determine the accuracy of these stability ideas.

6. The analysis of Brener et al.[15] for crystal growth in a channel, as well as arguments by Ungar and Brown[8], again suggest the existence of a branch of steady state solutions in DS other than the ST branch. The analytic mapping of the $p \to 0$ DS equations to the ST problem breaks down near the LCS line $\zeta_t \approx 0$. Thus we argue that the matching solutions lie on a different, and physically irrelevant, branch of steady state solutions for DS.

COMPARISON OF PREDICTIONS FOR GROOVE WIDTH, WAVELENGTH AND TIP POSITION

In this section, we illustrate the points 1, 2, 4 and 5 made above in more detail.

Let us first make the comparison of our results with experimental observations more quantitative with the aid of the data of de Cheveigné et al.[16] shown in Fig. 1. In this figure, the wavelength of perturbations about the planar interface that are neutrally stable (i.e. neither grow nor decay) is indicated by a solid line. Curves for two different temperature gradients are shown. The minimum threshold velocity V_c at which the planar interface first goes unstable is denoted by the square symbol at the bottom of the curves. Perturbations about the planar interface with wavelengths in the wide band between these two solid lines are linearly unstable; we therefore refer to this region as the planar instability band. The selected cells for a given pulling velocity in the experiments of de Cheveigné et al.[16] have wavelengths that lie within this planar instability band in the narrow dashed region whose wavelength is a factor 3 ~ 5 larger than the neutral stability value. This is what is typically observed.

A quantitative measure of the importance of surface tension effects is the parameter σ, defined as

$$\sigma \equiv \left[\frac{\lambda_s^0}{\pi\lambda}\right]^2 , \qquad (1)$$

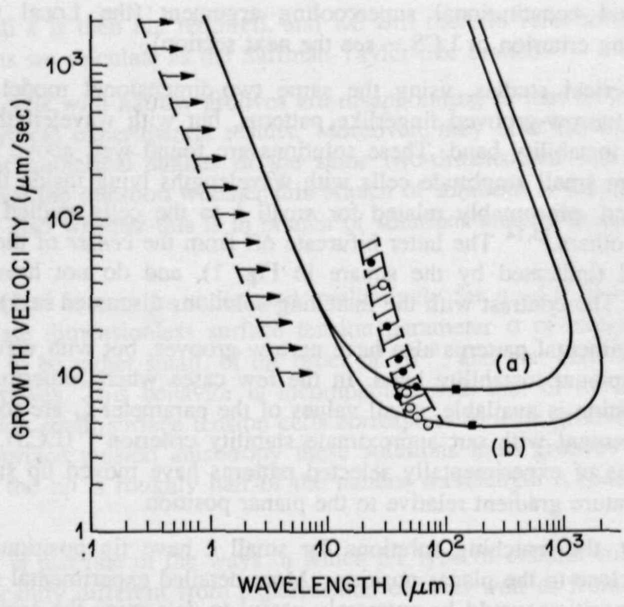

FIGURE 1

Plot of the growth velocity versus wavelength for the experiments of de Cheveigné et al.[16]. The solid line marks the neutral stability wavelength as a function of the growth velocity for two values of the thermal gradient G. Experiments for (a) $G = 120\,°C/cm$, full circles and (b) $G = 70\,°C/cm$, open circles lie in the narrow shaded band. The dashed line indicates the minimum deep cell wavelength calculated in this paper, and the arrows illustrate that the matched asymptotic expansion employed in this paper is an expansion towards larger wavelength.

where

$$\lambda_s^0 = 2\pi \left[\frac{\nu d_0 \ell_D}{\nu - 1} \right]^{1/2} \tag{2}$$

is the $k \to 0$ limit of the smallest neutral stability wavelength λ_s. Here d_0 is the usual capillary length, $\ell_D = D / V$ the diffusion length and $\nu = \ell_T / \ell_D$ the ratio of the thermal length ℓ_T and the diffusion length. (Throughout this paper, we will use the notation of Ref. 7) Since $k \approx 0.16$ for de Cheveigné's data[16] and since the k dependence of λ_s is rather weak for such small values of k, we may ignore the difference between λ_s and λ_s^0 and estimate $\lambda_s^0 / \lambda = 0.2\text{~}0.25$ for the data of Fig. 1. Thus $\sigma \approx 0.006$ in this experiment. On the other hand, as the arrows in Fig. 1 indicate, the narrow groove solutions that we can calculate perturbatively using matched asymptotic expansion methods have $\lambda_s^0 / \lambda = 2 \sim 3$, so that for these $\sigma = O(1)$.

A quantity that is explicitly predicted from the mapping onto the ST problem as well as from our matched asymptotic expansion approach for finite p, is the tip position z_t of the cells. We take the convention that the cells grow in the $+z$ direction and that the planar position is at $z = 0$. According to our calculations, the p-dependence of the ratio z_t / ℓ_T of cells on the ST branch is quite weak, and we will therefore approximate $z_t(p) / \ell_T$ by $z_t(p \to 0) / \ell_T$ as predicted by the ST analogy. The results are plotted in Fig. 2 as a function of ν for several values of the relative groove with ε. As Fig. 2 illustrates, the larger the groove width ε, the more the cells are predicted to move up for fixed ν. The data points in Fig. 2 refer to experimental measurements of Esaka and Kurz[17] on succinonitrile-acetone mixtures with $k = 0.1$. Clearly, the experimental cells have moved up in the temperature gradient more than any of the ST solutions predicts they should in the absence of crystalline anisotropy. Moreover, the solutions of the ST branch that come closest to the experimental data points are those corresponding to *wide* grooves ($\varepsilon \to 1/2$). Therefore, both Fig. 1 and Fig. 2 give evidence that the solutions on the ST branch have much wider grooves than one commonly observes.

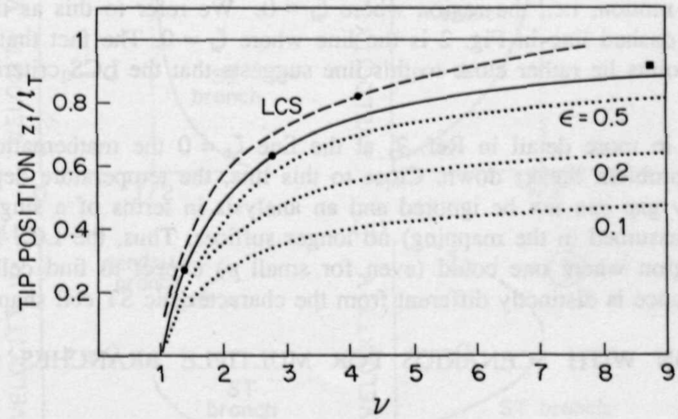

FIGURE 2

Plot of the tip position z_t/ℓ_T versus ν. The dashed line corresponds to the LCS line given by $\zeta_t = 0$ or $z_t/\ell_T = (1-1/\nu) / (1-k)$ for $k=0.1$. The full line gives the $k \to 0$ limit of the LCS line, $z_t/\ell_T = 1 - 1/\nu$, which is a relation first derived by Brody and Flemings (See Ref.7). The dotted lines are the expression for $z_t(p) / \ell_T$ as calculated in Ref. 3 and 4. The solid dot at $\nu=4.2$ and the square dot at $\nu=8.7$ represent two data points obtained and published by Esaka and Kurz,[17] while the data points for $\nu=1.3$ and $\nu=4.2$ refer to measurements by the same authors reported by Billia et al.[17]

The dashed line in Fig. 2 is labeled LCS. This stands for Local Constitutional Supercooling, an approximate stability and selection criterion for cells that we have advanced recently.[11] This criterion is a straightforward extension of the classical constitutional supercooling theory for the stability of a plane, whose basic idea is that in the steady state we can apply the concept of local thermodynamic equilibrium to the *melt just in front* of the moving planar interface. The interface is supposed to remain stable as long as the impurity concentration in the melt just in front lies in the stable single phase region as determined from the phase diagram. As is well-known, this idea predicts that a planar interface remains stable as long as $\nu \leq 1$. For small values of k, this criterion is quite accurate.

These constitutional supercooling arguments seem nearly as plausible when applied *locally* to the melt in front of the tips of certain (non-planar) patterns, provided that the curvature corrections are small (i.e. $\sigma << 1$), and that k is reasonably small, e.g., $k \leq 0.2$. In contrast to the plane, whose position in the gradient is fixed through conservation, cells can regain stability by moving up in the temperature gradient. A simple analysis based on this idea leads to the LCS prediction that cells restabilize at a position z_t such that

$$\zeta_t \equiv (\nu-1)-(1-k)z_t/\ell_D \leq 0 \ . \tag{3}$$

Equation (3) confirms that the tip position z_t of stable patterns with $\nu>1$ must move *up* in the cell, relative to the planar position at $z=0$. It is natural to conjecture that the operating point in real experiments should be close to the one that requires the least forward motion, i.e., the region where $\zeta_t \approx 0$. We refer to this as the LCS criterion.[11] The dashed line in Fig. 2 is the line where $\zeta_t = 0$. The fact that the experimental data points lie rather close to this line suggests that the LCS criterion is rather accurate.

As discussed in more detail in Ref. 7, at the line $\zeta_t = 0$ the mathematical mapping onto the ST problem breaks down. Close to this line, the temperature dependence of the miscibility gap can not be ignored and an analysis in terms of a single matching region (as is assumed in the mapping) no longer suffices. Thus, the LCS line roughly marks the region where one could (even for small p) expect to find cellular shapes whose appearance is distinctly different from the characteristic ST cell shape.

COMPARISON WITH SCENARIOS FOR MULTIPLE BRANCHES OF SOLUTIONS

In this section, we discuss our results in the light of other work on bifurcations and multiplicity of steady state cellular patterns (points 3 and 6 above). For a detailed discussion of the differences and similarities with the cell shapes calculate numerically by several groups we refer to Ref. 7.

In the work of Dombre and Hakim[2] and our extension thereof, cellular shapes with narrow grooves are calculated by matching an extension of the Scheil equation for the grooves to profiles that resemble finite amplitude solutions. However, the finite amplitude solutions that are used in this procedure are most probably physically irrelevant, since they lie *outside* the planar instability band. Moreover, these solutions on the ST branch are found to exist for *any* value of ν above the threshold ν_c, rather than a finite distance above threshold.[7] We believe that other possible branches of cells with grooves could be thought of as arising from joining up grooves to finite amplitude cells that lie within the planar instability band, even though the idea of a single matching region need not necessarily hold true. Evidence for the latter type of finite amplitude cells is given by the analytical work of Langer,[13] Sivashinsky[12] and Kurtze[14] as well as by the numerical work of Ungar and Brown.[8] Note in this regard that in contrast to the solutions our matching method finds, the cellular profiles discussed by

Sivashinsky[12] and Kurtze[14] for small k bifurcate off from the *center* of the planer instability band (indicated by the square in Fig. 1) and do *not* have a regular k expansion. Presumably some members of this latter finite amplitude branch of solutions develop deep grooves as ν is increased. Indeed, to our knowledge most numerical calculations[8–10] find deep cells by evolving continuously from small amplitude cells near the center of the planar instability band (near the square in Fig. 1).

Our conclusion that there is possibly another branch of steady state cellular solutions which is the physically relevant one for DS appear to be in line with the arguments of Brener et al.[15] discussed at this summerschool. These authors studied crystal growth in a channel, which corresponds to the $\ell_T \to \infty$ limit of DS. Not surprisingly, for

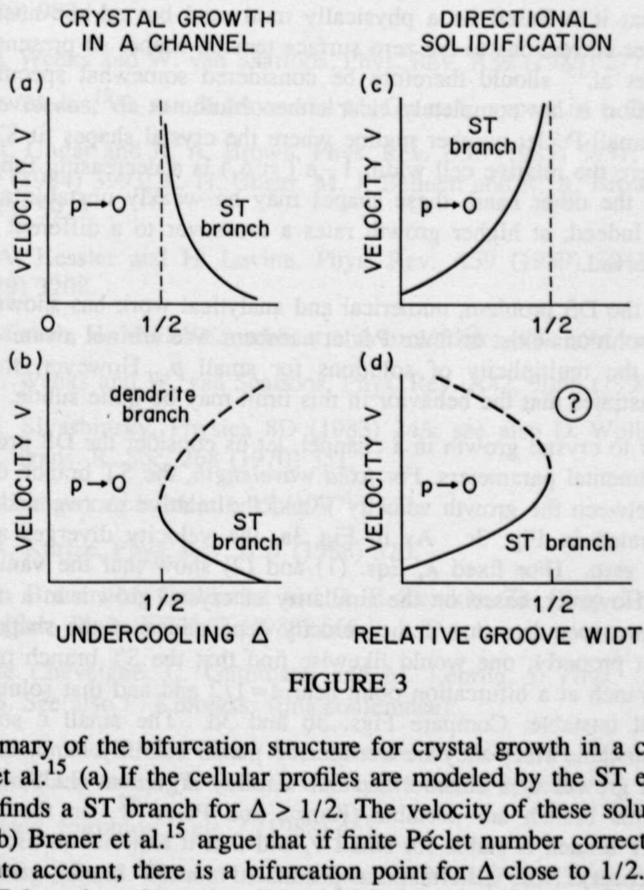

FIGURE 3

(a),(b) Summary of the bifurcation structure for crystal growth in a channel as argued by Brener et al.[15] (a) If the cellular profiles are modeled by the ST expression for the shape, one finds a ST branch for $\Delta > 1/2$. The velocity of these solutions diverges as $\Delta \to 1/2$. (b) Brener et al.[15] argue that if finite Péclet number corrections to the shape are taken into account, there is a bifurcation point for Δ close to 1/2. From this point, both the ST branch and another ''dendritic'' branch bifurcate. Solutions on the ST branch are unstable, and those on the dendritic branch stable. (c), (d) Possible bifurcation diagram for directional solidification for small p, based on the analogous conjecture for crystal growth in a channel. (c) The velocity V diverges as the relative groove width approaches 1/2. (d) In analogy with (b), it is possible that the ST branch merges with another cellular or dendrite branch near $\varepsilon = 1/2$ and that solutions on the ST branch are unstable.

small p, there is again a branch of Saffman-Taylor like solutions. From the mapping to the ST problem, it follows that these solutions exist only for dimensionless undercoolings $\Delta > 1/2$, and that the velocity is a *decreasing* function of Δ. This is illustrated in Fig. 3a. The decrease of V with increasing undercooling is, of course, counterintuitive, and both analytical[18] and numerical[19] work indicate that the solutions in this branch are *unstable*. Note also that on this ST branch, the velocity V becomes arbitrarily large as $\Delta \rightarrow 1/2$. For fixed channel width, this means that finite Péclet number corrections to the shape will become more and more important as $\Delta \rightarrow 1/2$. Motivated by this observation, Brener et al.[15] argue that in the absence of surface tension anisotropy, the ST branch ends at a bifurcation point near $\Delta = 1/2$. See Fig. 3b. Another "dendrite-like" branch bifurcates from this point as well, and it is this second branch of solutions that is believed to be stable[15,20].

Although the results of the simulations which Hunt discussed at the school[20] give some support for this scenario, we emphasize that the analysis of Brener et al.[15] is non-rigorous in that it is based on a physically motivated but ad-hoc ansatz for the finite Péclet number corrections to the zero surface tension shape. At present, the arguments of Brener et al.[15] should therefore be considered somewhat speculative. The experimental situation is not completely clear either. Molho et al.[21] observe that there is a low-velocity small-Péclet number regime where the crystal shapes fit ST solutions very well and where the relative cell width $1-\varepsilon$ ($=\Delta$) is a decreasing function of V, as in Fig. 3a. On the other hand, these shapes may be weakly unstable as expected theoretically[18,19]. Indeed, at higher growth rates a crossover to a different high velocity regime is observed.

Returning now to the DS problem, numerical and analytical work has shown that multiple branches of solutions exist at finite Péclet numbers. We are not aware of any systematic study of the multiplicity of solutions for small p. However, the work of Brener et al.[15] illustrates that the behavior in this limit may be quite subtle.

Indeed, in analogy to crystal growth in a channel, let us consider the DS problem for a fixed set of experimental parameters. For *fixed wavelength*, the ST branch of solutions gives a relation between the growth velocity V and the relative groove width ε whose behavior is illustrated in Fig. 3c. As in Fig 3a, the velocity diverges as $\varepsilon \rightarrow 1/2$, where σ tends to zero. [For fixed λ, Eqs. (1) and (2) show that the vanishing of σ implies $V \rightarrow \infty$.] However, based on the similarity of crystal growth in a channel and the DS problem, we speculate that if the velocity dependence of the shape would be taken into account properly, one would likewise find that the ST branch merges with another cellular branch at a bifurcation point near $\varepsilon = 1/2$ and and that solutions on the ST branch are all unstable. Compare Figs. 3b and 3d. The small ε solutions are almost certainly unstable since they lie outside the planar stability band, and both the analogy to crystal growth in a channel and our stability argument (LCS) suggest that all members of this branch are unstable (Karma and Pelcé [22], on the other hand, believe that the ST branch is stable for small ν, and that it exhibits an oscillatory instability as ν increases). Note also that the postulated non-ST branch corresponds to much larger velocities at small ε than the ST branch. For fixed wavelength λ, (1) shows that this conjectures branch is likely to have small groove width ε *and* small σ – precisely the characteristics that distinguish experimental cells form those on the ST branch! We hope that future analysis of the ideas of Brener et al.[15] will establish whether this scenario is correct.

CONCLUSION

In conclusion, we believe that there are serious indications that the Saffman-Taylor branch of cellular solutions is not the relevant one for DS. This issue is an important assumption that this branch does in fact correspond to the one that one observes in experiments. The resolution of this question will have important implications for our understanding of DS as well as crystal growth in a channel.

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